Dkt. 66194-Z CCD

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appellants: Paul Vincent Evans,
Theodor Rottwinkel and
Jeremy Mark Brown

Serial No. : 10/726,181 — Group Art Unit 1742

Filed : December 1, 2003 — Examiner J. Combs-Morillo

For : ALUMINIUM ALLOY SHEET

BRIEF ON APPEAL

This is appellants' Brief under 37 C.F.R. §41.37 in support of their appeal to the Board of Patent Appeals and Interferences under 37 C.F.R. §41.31 from the final rejection of claims 19 - 26 of the above-identified application. Attached hereto is a check in payment of the requisite fee of $500.00 under 37 C.F.R. §41.20(b)(2) for filing a brief in support of an appeal. If any additional fee is required, please charge the amount of such fee to Deposit Account No. 03-3125.

(I) Real Party in Interest

The real party in interest is Novelis, Inc., a corporation having a place of business at 70 York Street, Suite 1510, Toronto, Ontario M5J 1S9, Canada, assignee of the above-identified application by virtue of an assignment (recorded December 22, 2005, at Reel 016934, Frame 0600) from Alcan International Limited, the previous assignee of the above-identified application by virtue of an assignment (recorded February 26, 2002, at Reel 012651, Frame 0403) from the named appellants.

(ii) Related Appeals and Interferences

None.

(iii) Status of Claims

Claims 19 through 26 were presented in a Preliminary Amendment filed December 1, 2003, concurrently with the filing of the above-identified application, which is a division of appellants' U.S. patent application Serial No. 09/980,886 (now abandoned) filed February 8, 2002, as the U.S. national stage of international application No. PCT/GB00/02026 filed May 26, 2000.[1] No claim has been amended or cancelled. Claims 19, 20, 21, 22, 23, 24, 25 and 26 are all pending and all finally rejected. The claims appealed are all the pending claims, viz., claims 19, 20, 21, 22, 23, 24, 25 and 26. NOTE: Claim 19 is independent; all the other claims are dependent. The only grounds of rejection in the final Office Action are rejections on prior art under 35 U.S.C. §103(a).

(iv) Status of Amendments

No amendment was filed subsequent to the final rejection (Office Action dated January 25, 2005) from which the present appeal is taken.

(v) Summary of Claimed Subject Matter

In the following summary, all page and line numbers refer to appellants' specification. There are no drawings.

The invention defined by the claims on appeal embraces a method of producing an aluminum lithographic sheet (lithographic plate support) (p. 1, lines 31-32; original claim 13, p. 14, lines 19-20) from direct chill (DC) cast ingot. In such sheet, it is important to achieve a satisfactory graining response free of surface defects of various kinds when the sheet is subjected to electrograining (p. 1, lines 5-8). Specifically, a streaky electrograined surface is to be avoided (p. 3, lines 22-23; p. 4, lines 8-9 and 16-18), as is the presence of a region containing Al_mFe phase, known as a "fir tree zone," at scalp depth in the cast ingot from which the sheet is produced (p. 6, lines 1-13).

It is known that the ingot must be grain refined in order for the Al_mFe phase (fir tree structure) to appear (p. 6, lines 16-17). Grain refining results from the addition of grain refiners, e.g. Ti and B, to the alloy of which the ingot is cast (p. 7, lines 3-24). To avoid fir tree structure at scalp depth in a grain refined aluminum alloy, the maximum possible ingot-casting speeds are inconveniently slow (p. 6, lines 17-19).

[1]The Preliminary Amendment cancelled all the claims (1 - 18) originally presented in the parent application.

Nevertheless, in conventional practice, aluminum sheet for lithographic plate support has been grain refined (p. 5, lines 31-32), by addition of grain refiners (p. 4, lines 12-15), to generate a uniform distribution of equiaxed small, randomly oriented grains at the scalp depth (p. 4, lines 1-2) as well as to avoid excessive microporosity (p. 4, lines 3-11). The absence of grain refiner results in production of very coarse grains that have previously been thought to lead to the formation of a defect (streaking) during electrograining of the final gauge sheet (p. 4, lines 12-18).

The lithographic sheet-producing method of the present invention, as defined in claim 19 on appeal, comprises providing a molten body (p. 1, line 6; p. 9, lines 13-16) of an aluminum alloy of the following composition in wt%:

Si	0.05 - 0.20
Fe	0.25 - 0.40
Others up to 0.05 each and up to 0.15 total	
Al	balance

(p. 2, lines 1-6; p. 10, lines 6-14) wherein the aluminum alloy melt is prepared ***without the addition of grain refiners*** (p. 2, lines 1-6; p. 6, lines 23-24; p. 7, lines 3-4 and 9-13); optionally degassing the molten body (p. 8, lines 6-15); direct chill (DC) casting the molten body to form a cast ingot (p. 9, lines 13-17); rolling the ingot to sheet (p. 1, line 6); and electrograining the rolled sheet (p. 1, line 8; p. 2, line 30 - p. 3, line 1).

In particular embodiments of the claimed method, the cast ingot has a hydrogen content of not more than 0.25ml/100g (claim 20; p. 7, lines 27-28), the Fe/Si ratio is from 2.5 to 5.5 (claim 21; p. 3, lines 13-15; p. 10, line 15), the DC casting speed is >60mm/min (claim 22; p. 9, lines 20-21), the cast ingot contains grains of a length >500 μm (claim 23; p. 4, lines 30-32), the cast ingot contains substantially no Al_mFe (claim 24; p. 6, lines 13-14), the iron in solution is 0.0018 to 0.0051 wt% (claim 25; original claim 12, p. 14, lines 16-17), and the electrolyte used for electrograining is nitric acid (claim 26; p. 10, line 1-2).

Surprisingly, in relation to what was previously thought, the present inventors have found that the coarse grain structure resulting from absence of grain refiner is not in itself detrimental to final gauge electrograining response (p. 4, lines 18-20). The absence of grain refiner avoids the detrimental fir tree structure at scalp depth and thus enables ingots (for production of lithographic plates) to be DC cast at higher casting speeds than have heretofore generally been possible (p. 6, lines 20-22). That is to say, with grain refiner absent (in accordance with the invention), casting speed is not critical, and to achieve high throughput and low costs, casting speed may be as fast as possible, with a maximum limit imposed by risk of run-out and safety and practical details rather than by metallurgical considerations (p. 9, lines 16-21).

(vi) Grounds of Rejection to be Reviewed on Appeal

The sole ground of rejection presented for review on this appeal is as follows:

That claims 19 - 26 are unpatentable under 35 U.S.C. §103(a) over S. Brusethaug et al., "The Effect of Process Parameters on the Fir-Tree Structure in DC-Cast Rolling Ingots," Special print of the documentation of 8th ILMT, 1987, Leoben-Vienna, pp. 472-76 (Brusethaug et al.) in view of U.S. patent No. 5,711,827 (Sawada et al. '827).

(vii) Argument

Rejection under 35 U.S.C. §103(a) as unpatentable over Brusethaug et al. in view of Sawada et al. '827

It is submitted that the rejection of claims 19 - 26 on Brusethaug et al. in view of Sawada et al. '827 is in error in that, contrary to the Examiner's contention, it would not have been obvious to a person of ordinary skill in the art, from the applied references, considered together, to produce an aluminum lithographic sheet by DC casting an alloy of the composition of appellants' claims to form an ingot, rolling the ingot into sheet, and electrograining the sheet, without the addition of grain refiners to the aluminum alloy being cast. Stated in other words, it would not have been obvious from the references that if one were to take a non-grain-refined DC ingot, roll it down to final gauge and then electrograin it, a product could be obtained having no streaky grains.

Appellants' claim 19 recites, in pertinent part, the steps of (a) providing a molten body of aluminum alloy of specified composition (0.05-0.20 wt% Si, 0.25-0.40 wt% Fe) "wherein the aluminum alloy melt is prepared without the addition of grain refiners," (c) DC casting the molten body to form a cast ingot, (d) rolling the ingot into sheet, and (e) electrograining the rolled sheet. The provision of this combination of composition features and process steps, including the electrograining step, *without the addition of grain refiners*, is not described in the prior art.

Stated with reference to the rejection of claim 19, the Examiner's position is that Brusethaug et al., directed to the making of offset plates without defects such as fir tree structures, describes a DC-cast sheet ingot of grain-refiner-free Al-Fe-Si alloy, and that it would have been obvious to apply to this ingot the rolling and electrograining steps described by Sawada et al. '827 for producing Al-Fe-Si alloy printing plates from DC-cast ingots.

Appellants respectfully submit that this asserted combination of references would not have been obvious, because neither Brusethaug et al. nor Sawada et al. '827 teaches or suggests that ingots without grain refiner could be used to make lithographic sheet.

The only grain-refiner-free ingot mentioned by Brusethaug et al. is the single ingot described at the top of the second column on p. 473 as having "no grain refinement." This ingot, and another of the same dimensions and composition (0.26 wt% Fe, 0.13 wt% Si) but containing 10/1000% Ti (grain refiner), were cast under identical conditions to investigate "The effect of grain refinement on the formation of fir tree zones." The result of this investigation, as reported by Brusethaug et al., was that "The fir-tree zone was only observed in the ingot with a grain refiner addition."

However, Brusethaug et al. clearly shows that two factors were considered important to avoid: non-uniform grain and the fir-tree structure. The mere elimination of fir-tree structure, without also overcoming the problem of non-uniform grain structure, would not have been deemed enough to achieve a satisfactory and usable product, in the state of the art as exemplified by Brusethaug et al.

As the Introduction to Brusethaug et al. states (p. 472, first column), the publication reports "work on the appearance of the fir-tree structure and its dependence upon casting conditions and alloy composition." The Introduction further explains that

> "Certain products like offset plates . . . require a homogenous surface . . . .
>
> "A typical defect . . . is structural streaking . . . which is unacceptable in offset . . . quality. Structural streaking can be the result of a non uniform grain structure (1), segregation or local differences in the primary constituents within the aluminium matrix – the so-called fir-tree structure (2-6)."

Reference (1) is a textbook by D. Altenpohl, *Aluminum Viewed from Within* (Dusseldorf: Aluminium-Verlag, 1982) of which a copy of the passage (pp. 146-47) cited by Brusethaug et al. is of record in the file of the present application. Discussing "Causes of Streaking or Streak-Free Structure After Forming," Altenpohl states that streaks may be "due to large as-cast grains," that "the streaking from large as-cast grains even after severe cold work is not difficult to understand," and that "It is important to avoid coarse grain in all processing steps." Hence, the above-quoted Introduction of Brusethaug et al. effectively teaches that undesirable structural streaking can result (*inter alia*) from either coarse grains or fir-tree structure.

From this, a person of ordinary skill in the art could only conclude that *both* coarse grains *and* fir-tree structure must be avoided in order to prevent structural streaking. Thus, while the

investigation reported by Brusethaug et al. is a study of fir-tree structure, aiming "to eliminate the fir-tree structure, or at least to keep the zone width consistently smaller than the scalping depth," there is no implication in the reference that such control of fir-tree structure is a sufficient condition for prevention of structural streaking and consequent production of satisfactory printing plates.

Reverting again to the description of the single grain-refiner-free ingot mentioned by Brusethaug et al. (p. 473, second column, top), appellants note that immediately after reporting the absence of a fir-tree zone in this ingot, the reference states that "Casting speed and Fe/Si ratio are clearly dominating parameters regarding the fir-tree zone formation." Inferentially, then, the grain-refiner-free ingot did not itself represent a useful or practical solution to the fir-tree zone problem but was merely an investigatory test of one of multiple factors. The Brusethaug et al. publication supplies no thought or suggestion of departing from the standard practice of including grain refiner in any DC-cast ingot that is to be rolled and electrograined for production of printing plates. Rather, Brusethaug et al., taken as a whole, reinforces the view then standard in the art that the presence of grain refiner is essential. So much is evident from the explicit but unambiguous teaching, in the introductory portion of the reference (and specifically in the citation of Altenpohl), that to prevent structural streaking, coarse grains must be avoided – i.e., that grain refiner (to control grain size) is essential.

Certainly there is nothing in Sawada et al. '827 to suggest such a departure from standard practice. Sawada et al. is absolutely silent regarding any modification or omission of the conventional procedure of adding grain refiner, nor does Sawada et al. remotely intimate that, contrary to what was generally thought, it would or might not be necessary to avoid the coarse grain structure that results from the absence of grain refiner in order to achieve a satisfactory final gauge electrograining response in the production of lithographic sheet.

An artisan of ordinary skill, therefore, considering the two references together, would not have been led to suppose that the investigatory grain-refiner-free ingot of Brusethaug et al. could produce satisfactory printing plates if subjected to the process steps of Sawada et al. '827. In other words, the artisan would not find in the two references any motivation to combine them as the Examiner has asserted.

In summary: it is the Examiner's position that (1) Brusethaug et al. shows a DC cast ingot free of grain refiner, in which fir tree structure was not observed, (2) Sawada et al. '827 teaches process steps of producing a lithographic sheet, including rolling a DC cast ingot and electrograining, and (3) it would have been obvious to perform the steps of Sawada et al. '827 on the grain-refiner-free ingot of Brusethaug et al., because the latter ingot is a rolling ingot and both references are drawn to printing plates. But assuming *arguendo* the correctness of the Examiner's assertions (1) and (2), nevertheless it would not have been obvious to combine them as proposed in the Examiner's

assertion (3), because Brusethaug et al. implicitly teaches away from any notion that the grain-refiner-free ingot, though lacking fir tree structure, would be suitable or capable of being formed into satisfactory lithographic sheet, and Sawada et al. '827 is silent in this regard.

That is, Brusethaug et al. expressly states that unacceptable structural streaking can result either from non uniform grain structure or from fir tree structure; and while the reference reports that one cause of streaking (fir tree structure) was not present when grain refiner was omitted, a person of ordinary skill in the art would have assumed that the absence of grain refiner would lead to the other cause of streaking (very coarse grains), hence, that satisfactory lithographic sheet would not be obtained if the investigational grain-refiner-free ingot were subjected to rolling and electrograining. Sawada et al. '827 contains no disclosure to suggest any different conclusion.

Coarse grains are indeed observed when grain refiner is not added. As appellants' specification sets forth, in their claimed method the ingot may have grains longer than 500 μm (p. 4, lines 29-30). Appellants have discovered that, contrary to what was thought before, this does not prevent the production of satisfactory lithographic sheet, but without their discovery, it would not have been obvious to perform the steps of Sawada et al. '827 on the grain-refiner-free ingot of Brusethaug et al. because the ordinarily skilled artisan would have no reason to expect that an acceptable lithographic sheet product would result.

It follows that claim 19, in reciting that the alloy melt is prepared "without the addition of grain refiner" and is subjected to steps of DC casting, rolling and electrograining, presents an unobvious and patentable distinction over Brusethaug et al., Sawada et al. '827 and any proper combination thereof. The ability of a grain-refiner-free alloy, so processed, to achieve an acceptable lithographic sheet product, in itself represents a surprising new result, and affords the important economic advantage of enabling a beneficial increase in speed of casting the DC ingot from which the sheet is formed, without causing the deleterious fir tree structure.

It may also be noted that the subjection of the specific grain-refiner-free alloy of the Brusethaug et al. test ingot (containing 0.26 wt.% Fe, see Brusethaug et al., p. 473, right column, line 6) to the process of Sawada et al. '827 would be contrary to the express teaching of Sawada et al. '827 that the alloy should have a content of not more than 0.20 wt.% Fe (see Sawada et al., col. 5, lines 22-26), and would therefore be unobvious on that ground as well. If the alloy were modified to meet the Sawada et al. upper limit of Fe content, it would be below appellants' claimed range of 0.25-0.40 wt.% Fe.

Claims 20 - 26 are submitted to distinguish in like manner over any proper combination of Brusethaug et al. and Sawada et al. '827 by virtue of their dependence on claim 19.

(viii) Claims Appendix

A copy of the claims on appeal is set forth in an Appendix immediately following the conclusion and signature page.

(ix) Evidence Appendix

Copies of the following references are submitted in an Evidence Appendix immediately following the Claims Appendix:

1. S. Brusethaug et al., "The Effect of Process Parameters on the Fir-Tree Structure in DC-Cast Rolling Ingots," Special print of the documentation of 8th ILMT, 1987, Leoben-Vienna, pp. 472-76.
2. U.S. patent No. 5,711,827, Sawada et al.
3. D. Altenpohl, *Aluminum Viewed from Within* (Dusseldorf: Aluminium-Verlag, 1982), pp. 146-47.

(x) Related Proceedings Appendix

None.

Conclusion

For the foregoing reasons, it is respectfully requested that the decision of the Examiner rejecting claims 19 - 26 be reversed, and that the claims be allowed.

Respectfully,

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I hereby certify that this paper is being deposited this date with the U.S. Postal Service as first class mail addressed to Assistant Commissioner for Patents, Washington, D.C. 20231.

Christopher C. Dunham

Christopher C. Dunham
Reg. No. 22,031 Date JANUARY 3, 2006

Claims Appendix

19. A method of producing an aluminium lithographic sheet which method comprises:

a) providing a molten body of an aluminium alloy of composition in wt%:

Si	0.05 - 0.20
Fe	0.25 - 0.40
Others up to 0.05 each and up to 0.15 total	
Al	balance

wherein the aluminium alloy melt is prepared without the addition of grain refiners,

b) optionally degassing the molten body,

c) direct chill (DC) casting the molten body to form a cast ingot,

d) rolling the ingot to sheet,

e) electrograining the rolled sheet.

20. A method of producing an aluminium lithographic sheet according to claim 19 such that the cast ingot has a hydrogen content of not more than 0.25ml/100g.

21. A method of producing an aluminium lithographic sheet according to claim 19, wherein the Fe/Si ratio is from 2.5 to 5.5.

22. A method of producing an aluminium lithographic sheet according to claim 19, wherein the DC casting speed is >60mm/min.

23. A method of producing an aluminium lithographic sheet according to claim 19, wherein the cast ingot contains grains of a length >500μm.

24. A method of producing an aluminium lithographic sheet according to claim 19, wherein the cast ingot contains substantially no Al_mFe.

25. A method of producing an aluminium lithographic sheet according to claim 19, wherein the iron in solution is 0.0018 to 0.0051 wt%.

26. A method of producing an aluminium lithographic sheet according to claim 19, wherein the electrolyte used for electrograining is nitric acid.

Evidence Appendix

Copies of the following references are attached hereto:

1. S. Brusethaug et al., "The Effect of Process Parameters on the Fir-Tree Structure in DC-Cast Rolling Ingots," Special print of the documentation of 8th ILMT, 1987, Leoben-Vienna, pp. 472-76, cited by the Examiner in an Office Action dated May 5, 2004 (Brusethaug et al.).

2. U.S. patent No. 5,711,827, Sawada et al., cited by the Examiner in an Office Action dated May 5, 2004 (Sawada et al. '827).

3. D. Altenpohl, *Aluminum Viewed from Within* (Dusseldorf: Aluminium-Verlag, 1982), pp. 146-47, cited in Brusethaug et al., and submitted by appellants in a Reply under 37 C.F.R. §1.111 filed November 4, 2004, to which the final Office Action dated January 25, 2005, is responsive (Altenpohl).

OTR 18

THE EFFECT OF PROCESS PARAMETERS ON THE FIR-TREE STRUCTURE IN DC-CAST ROLLING INGOTS

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Introduction

Certain products like offset plates and anodized panels require a homogeneous surface. Beside careful attention to the fabrication process to avoid surface defects (grease marks, roll pick-up and mechanical damage) control of the microstructure of the sheet ingot is also required.

A typical defect arising from the microstructure is structural streaking (light and dark bands in the rolling direction) which is unacceptable in offset and anodizing quality. Structural streaking can be the result of a non uniform grain structure (1), segregation or local differences in the primary constituents within the aluminium matrix - the so-called fir-tree structure (2-6).

In this article, work on the appearance of the fir-tree structure and its dependence upon casting conditions and alloy composition is reported and a modified model for its formation is proposed.

Origin of the fir-tree structure

The types of primary constituents formed in a ingot cross section change, as a result of changing cooling rates, with distance from the ingot surface. After caustic etching of ingot slices fir-tree zones sometimes appear near the surface of unalloyed Al, figure 1. This has been explained by differences in etching characteristics resulting from a microstructure dominated by Al_mFe- primary constituents in the outer zone and Al_6Fe in the inner zone (2).

The boundary between the inner and outer zones is typically irregular. In extreme cases the inner zone takes the form of a fir-tree on vertical sections. Figure 1 is a horizontal section. In this article the outer zone will be referred to as the fir-tree zone. Small islands may appear within the fir-tree zone with an etching behaviour similar to the inner part of the ingot. In sheet ingots with small dimensions the zone may also appear in the centre of the ingot.

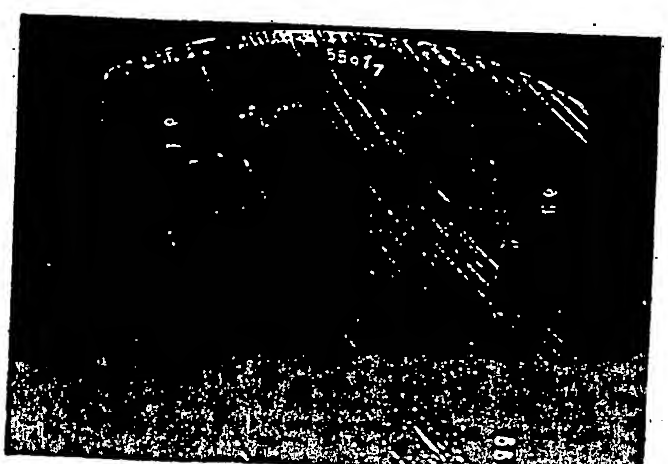

Fig. 1: Fir-tree zone

If the scalping of a sheet ingot is insufficient to entirely remove the fir-tree zone structural streaking appear on rolled material after etching. The aim is therefore to eliminate the fir-tree structure, or at least to keep the zone width consistently smaller than the scalping depth.

Experimental work and results

Control of fir-tree zone formation requires a knowledge of how the casting conditions and minor changes in chemical composition affect the width of the zone.

To reveal the effect of casting speed, secondary cooling, Fe and Si content and grain refining several full scale DC-casting trials of unalloyed sheet ingots (99.5 % Al) have been carried out. The ingots were cast either by the conventional downspout and float technique with a fibrefrax paper glued onto the upper part of the mould wall or by level pour with fibrefrax hot-top. Standard grain refinement used was 15/1000 % Ti added to furnace as waffle and 10/1000 % Ti added as AlSTi1B rod. Fir-tree zone widths were measured along 100 mm lengths on the long sides of the sheet ingots. The zones were traced on an image analyzer to obtain a mean and a maximum zone height.

Effect of Fe and Si content

The influence of Fe and Si content on the width of the fir-tree zone has been studied earlier by varying the Fe/Si ratio from 1/2 to 3 (7). It was found that a maximum fir-tree zone width occurs at a Fe/Si ratio of approximately 2. A Fe/Si-ratio near 1 has proven to be unacceptable with respect to grainability of the rolled sheet, and a possible way of avoiding fir-tree zones in this quality therefore seems to be to increase the Fe/Si ratio to at least 3.

The effect of increasing the Fe/Si ratio from 1.8 (0.29 % Fe, 0.16 % Si) to 5 (0.35 % Fe, 0.07 % Si) on the fir-tree zone width and type of primary constituents nucleated is shown in table 1. Samples were taken from sheet ingots with dimensions 600 x 1600 mm and particles dissolved from matrix by the butanol method (8,9) were identified in a JEOL-100CX TEM microscope (EDS, SADP). A correlation between Al_mFe as the dominant primary constituent and the appearance of the fir-tree zone was found.

Table 1: Primary constituents as a function of distance from surface (dominating primary constituent underlined). Sheet ingots cast with downspout and float technique.

Fe/Si	Observed primary constituents			Fir-tree zone	
	Distance from surface (mm)				
	10 and 20	150	300	Max.	Mean
5	[illegible]	AlFe	AlFe, AlFe	0	0
2	AlFe, AlFe, α-AlFeSi, AlFe	AlFe	AlFe, α-AlFeSi	36.4	25

Special print of the documentation of 8th ILMT, 1987, Leoben–Vienna

Effect of casting speed

Since the nucleation and growth of Al_mFe require high cooling rates formation of the fir-tree structure will be promoted by a high casting speed. Several casting experiments (10,11) with 600 mm wide sheet ingots, have shown that a reduction in casting speed from 75-80 to 60-65 mm/min reduces the problem of structural streaks in material with a Fe/Si ratio of 2 since the width of the fir-tree zone is then usually smaller than the scalping depth, table 2.

Table 2: Effect of casting speed on fir-tree zone width (casting technique: Downspout and float)

Fe (%)	Si (%)	Dimension (mm x mm)	Mould height (mm)	Casting speed (mm/min)	Mean fir-tree zone width (mm)
0.25	0.12	600 x 1600	35	65	55
0.25	0.12	600 x 1600	40	65	35
0.25	0.12	600 x 1600	45	65	50
0.25	0.12	600 x 1600	50	65	45
0.25	0.12	600 x 1600	65	65	65
0.25	0.12	600 x 1600	35	75	21
0.25	0.12	600 x 1600	45	75	22
0.25	0.12	600 x 1600	50	75	24
0.25	0.12	600 x 1600	65	75	19
0.22	0.11	600 x 1350	35	60	0
0.22	0.10	600 x 1350	70	80	0
0.22	0.11	600 x 1350	35	80	45
0.22	0.11	600 x 1350	50	80	27
0.26	0.13	600 x 1350	70	80	26

Sometimes, however, wide fir-tree zones also appear at low casting speeds suggesting that other factors in addition to casting speed and Fe and Si content influence the formation of fir-tree-zones.

Effect of secondary cooling

To see if a reduction in secondary cooling could alter the tendency to fir-tree zone formation trials with different amounts of cooling water were carried out at casting speeds of 65 and 80 mm/min. (13). The Fe/Si-ratio was 2 (0.26 % Fe, 0.13 % Si). The influence of changes in secondary cooling and casting speed on cooling rates/temperatures at different positions below the ingot surface was measured by thermocouple "harps" (12) cast into the ingot. Table 3 summarizes the results. At a casting speed of 80 mm/min a reduction of water rate reduces the cooling rate somewhat in the outer 20 mm of the sheet but only a minor decrease in fir-tree zone width is observed. The results for ingots cast at 65 mm/min are more unexpected and will be discussed later.

Table 3: Effect of secondary cooling

Drop [1]	Casting speed (mm/min)	Water rate (m3/h)	Fir-tree zone width (mm)		Cooling rate (°C/s) [3]			
					Dist. from surface (mm)			
			Mean	Max	10	20	40	80
1	65	30	45	55				
2	65	35	25	37				
2B [2]	65	35	15	16	2.2	4.3	4.0	1.9
3	65	17	20	34				
4	65	35	0	10		3.9	3.6	2.2
6	80	30	11	15	2.8	5.3	6.2	3.0
8	80	50	14	21	5.0	7.5	5.9	2.7
10	80	75	13	21	6.6	7.3	6.4	3.1

1) Level pour casting, 600 x 1600 mm sheet ingot, mould height 35 mm
2) No grain refiner rod addition
3) Temperature range 655 - 650 °C

Effect of grain refiner

The effect of grain refinement on the formation of fir tree zones in sheet ingots was investigated by casting 2 sheet ingots with dimensions 290 x 600 mm with a casting speed of 90 mm/min and Fe/Si=2 (0.26 % Fe, 0.13 % Si) with no grain refinement and with 10/1000 % Ti as AlTi5B1-rod respectively. The fir-tree zone was only observed in the ingot with a grain refiner addition.

Discussion

Casting speed, Fe/Si-ratio

Casting speed and Fe/Si ratio are clearly dominating parameters regarding the fir-tree zone formation. A casting speed of 60 - 65 mm/min is in most cases sufficiently low to prevent fir-tree zone formation at a Fe/Si ratio of 2 which normally results in the widest fir-tree zone.

The formation of the Al_mFe and Al_6Fe primary constituents causing the zone structure can be understood with the aid of a binary metastable Al-Fe phase diagram, figure 2a. Figure 2b shows schematically how the nucleation temperature of each phase might vary with cooling rate through the solidification interval. For a given cooling rate the phase with the highest nucleation temperature will nucleate first and start to grow. This has been used with some success to explain the influence of casting speed on fir-tree zone width. Figure 3 explains why the outer Al_mFe zone will be wider at a casting speed of 100 mm/min than at 60 mm/min according to this model. However this model for the formation of fir-tree structure does not explain why the zone sometimes also appeares at low casting speed, or why it very often extends through the shell zone cooling rate minimum. The model does not provide an explanation for the fir-tree shape or the coexistence of several different primary constituents near the ingot surface.

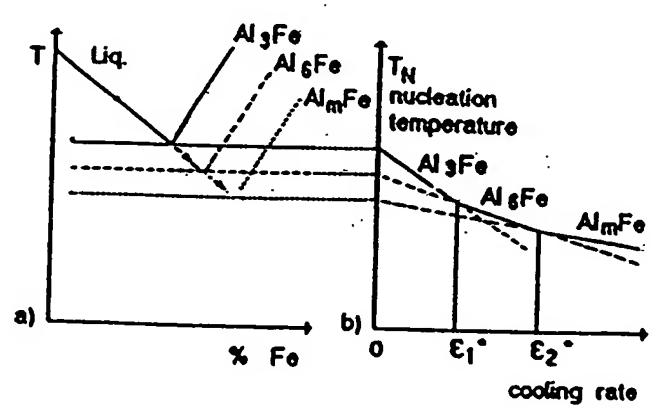

Fig. 2: a) Al-Fe phase diagram
b) Effect of cooling rate on the nucleation temperature

A Fe/Si ratio of 2 will result in the widest fir-tree zone. This is assumed to be because this ratio gives the lowest nucleation cooling rate threshold (ε_2) for Al_mFe, figure 3. An increase in the ratio seems to increase this threshold and Al_6Fe will instead be the dominant primary constituent. At low Fe/Si ratio α-AlFeSi will nucleate and be the dominant constituent at high cooling rates.

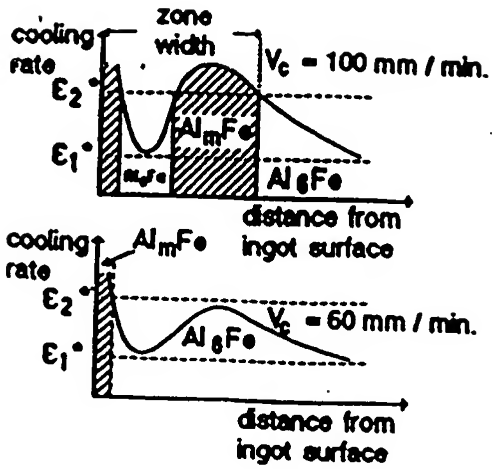

Fig. 3: The "conventional" model for fir-tree zone formation

Secondary cooling and grain refinement

At first sight the results for a casting speed of 65 mm/min in table 3 seem to be inconsistent; an increasing water flow rate reduces the fir-tree zone width. At 80 mm/min, however, a minor increase in fir-tree zone width with increasing water rate is detected.

A comparison of the results for casting speeds of 65 and 80 mm/min in table III shows that the fir-tree zone width decreases with increasing casting speed! To explain this unexpected result a closer look at chemical composition must be taken, expecially Ti and B contents, since grain refinement was shown to influence the zone width. A comparison between Ti and B contents and the mean zone width showed that the zone width is not directly related to the content of B or Ti, but a good correlation is obtained between the zone widths and B/Ti ratios, Figure 4. B/Ti-ratios at 80 mm/min casting speed were all about 0.05. Since it was also shown that grain refiner addition increases the tendency to fir-tree zone formation the results suggests that both grain refiner addition and the B/Ti ratio influences the fir-tree zone formation. The reason for this effect is however unclear.

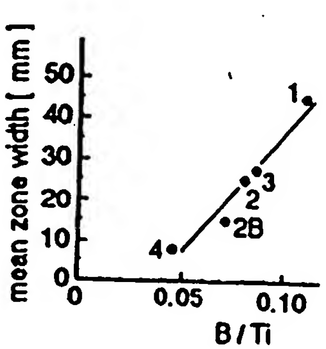

Fig. 4: The influence of B/Ti on the mean fir-tree zone width at 65 mm/min

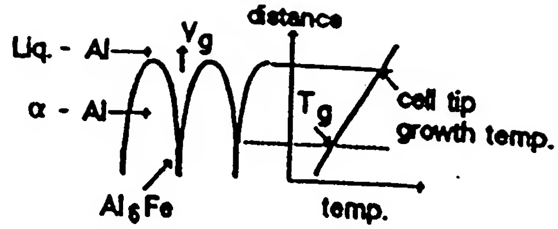

Fig. 5: Explaining the concept of growth. Temperature T_g is the growth temperature of Al_6Fe

A modified model for fir-tree formation (13)

The above model for fir-tree zone formation fails since it can only be used to predict which primary constituent will be nucleated during "isothermal cooling", i.e. when the temperature at any given time is uniform throughout the volume of solidifying liquid. This is not, however, the case during continuous casting or during the solidification of large castings.

Since temperature gradients exist the situation is more complicated. The growth temperatures of the competing phases must be taken into account. Consider the unidirectional growth of a cellular structure for example, as shown in Figure 5. Steady state growth is taking place in a constant temperature gradient. Under these conditions the growth front of the intermetallic phase will be located at a particular isotherm the temperature of which will depend on the imposed growth rate. Growth at a finite rate will require a certain undercooling below equilibrium (or metastable equilibrium) eutectic temperature in order to provide the necessary driving force to overcome curvature effects and to drive diffusion and the interface reaction.

The growth temperatures of Al_mFe and Al_6Fe might vary with growth rate as shown in figure 6.

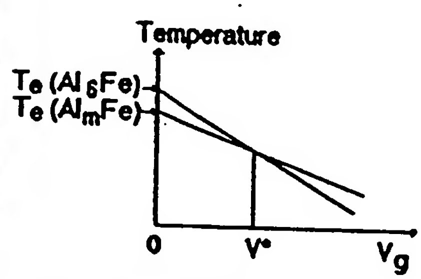

Fig. 6: The variation of T_g with growth rate

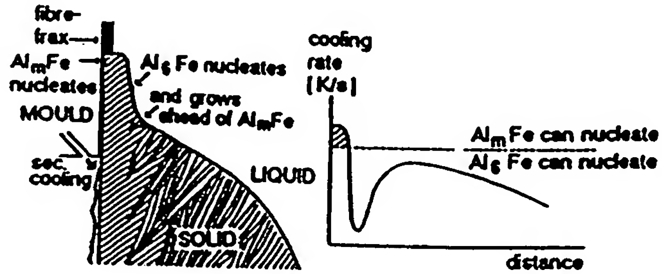

Fig. 7: Competive growth of Al_6Fe and Al_mFe leading to the formation of fir-tree structure

Consider now the situation during continuous casting. The ingot skin forms by the rapid quenching of the liquid against the water-cooled mould wall. Cooling rates are very high and we can expect Al_mFe to be the intermetallic phase that is first nucleated. The Al_mFe that has been formed between the aluminium dendrites moves down with the ingot, but continues to grow inwards. The intermetallic particles extend in a dendritic manner like fingers extending between the aluminium dendrites. The tips of the intermetallic "dendrites" will follow the isotherms corresponding to the instantaneous growth rate of the tips, as shown in Figure 5. The growth of the Al_mFe dendrites will continue until some other iron-rich phase nucleates and starts growing in front of the Al_mFe. Thus if the following conditions are satisfied:

$$T_N\ (Al_6Fe) > T_G(Al_mFe) \qquad (1)$$

and

$$T_G\ (Al_6Fe) > T_G(Al_mFe) \qquad (2)$$

Al_6Fe can nucleate and grow ahead of Al_mFe such that the growth of Al_mFe will be locally prevented. The above conditions are necessary, but not a sufficient criterion for the change from Al_mFe to Al_6Fe. There must also be suitable nucleation site for the Al_6Fe (this might be the primary aluminium dendrites, Al_mFe itself or some other impurity particles).

Conditions (1) and (2) above will be favoured by low cooling rates ahead of the Al_mFe dendrites, figure 7, and low growth rates, figure 6. Local growth rates and cooling rates are of course related to each other, but they may be influenced differently by casting speed and melt temperature for example.

The sequence of events leading to the formation of the characteristic fir-tree structure can now be explained as shown in figure 7. As soon as the cooling rate falls below some critical level, Al_6Fe can nucleate if a suitable nucleus is available. The most favorable conditions for the nucleation of the Al_6Fe are obtained at the point of inflection of the lip where the cooling rate is a minimum. Once Al_6Fe is nucleated it can grow at a higher temperature than Al_mFe ; provided the growth rate normal to the local isotherms is low enough. This enables the Al_6Fe dendrites to gradually encroach on the Al_mFe dendrites as shown in figure 7, leading to the characteristic fir-tree pattern. This sequence of events describes what happens at casting speeds up to 80 mm/min with large ingots. In other cases (higher speeds and/or smaller ingots) the second maximum in growth rate caused by secondary cooling, may be high enough to reverse the relative growth temperatures of Al_mFe and Al_6Fe such that Al_mFe encroaches on Al_6Fe and only isolated islands of Al_6Fe will be found near the shell zone, figure 9. Further towards the centre of ingot the cooling rate drops again, and the fir-tree structure develops near the centre of the ingot.

In small sheet ingots and in extrusion ingots a central zone is often visible in the centre of the ingot. This can again be explained since the growth rate in the centre of the ingot is as high as the casting speed. If the casting speed is higher than v* in figure 6 Al_mFe will be able to grow the whole length of the ingot in the central parts, figure 10.

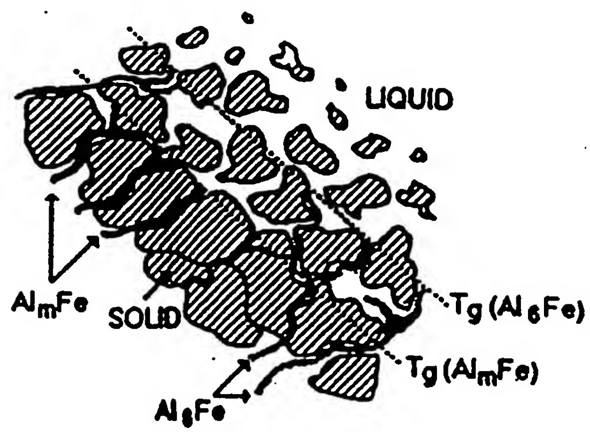

Fig. 8: A schematic view of the solidification front. $T_g\ (Al_6Fe)$ $T_g\ (Al_mFe)$

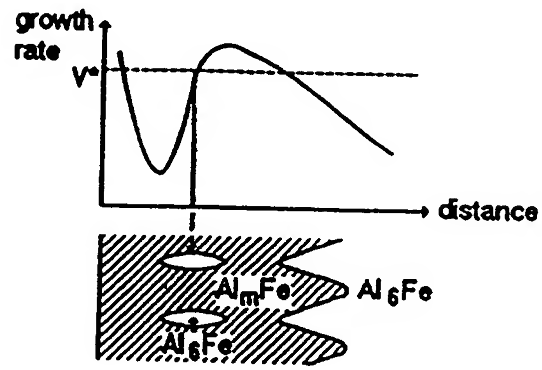

Fig. 9: Isolated islands of Al_6Fe

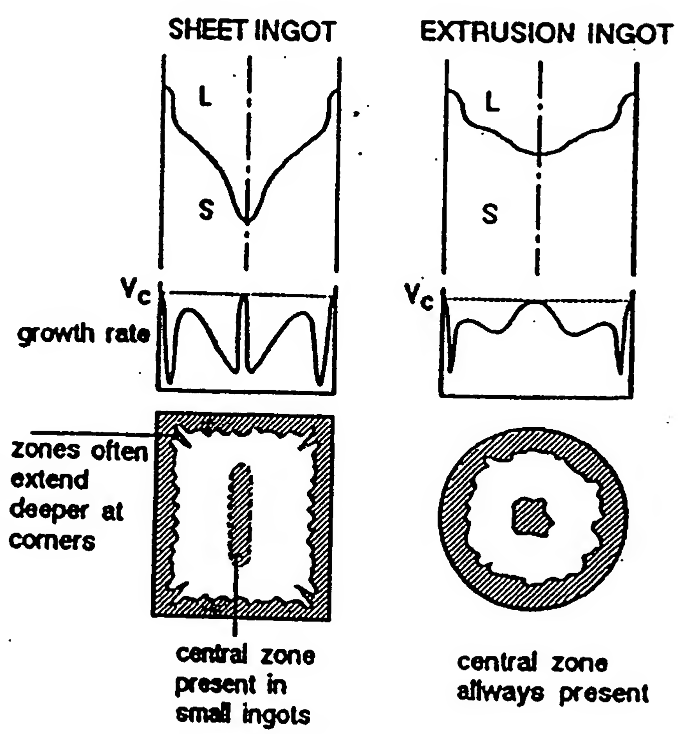

Fig. 10: Explaining the appearance of fir-tree zone in centre of ingots.

Acknowledgement

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References

1. D. Altenpohl: Aluminium viewed from within (1982) p. 146
2. N. Westengen: Paper presented at the 7. Int. Leichtmetalltagung Leoben-Wien, 22-26. Sept. 1981
3. I. Miki, H. Kosuge and K. Nagahama: J. Jap. Inst. L. Met. 32 (1975) 1-9
4. H. Kosuge and I. Mizukami: J. Jap. Inst. L. Met. 22 (1972) 437-444
5. H. Kosuge and I. Mizukami: J. Jap. Inst. L. Met. 25 (1975) 48-58
6. A. Asami, T. Tanaka and A. Hideno: J. Jap. Inst. L. Met. 28 (1978) 321-327
7. H. Westengen: Internal report, Hydro Aluminium, Sunndal Verk
8. C.J. Simensen and Spjelkavik: Frezenius Z. Anal. Chem., 300 (1980). p 177
9. C.J. Simensen, P. Fartum and A. Andersen: Frezenius Z. Anal. 319 (1984) 286-292
10. H. Westengen: Internal report, Hydro Aluminium, Sunndal Verk
11. E. Folmo: Internal report, Hydro Aluminium, Sunndal Verk
12. J.A. Bakken and T. Bergstrøm, Light Metals, 2, 883 (1986)
13. D. Porter: Internal report, Hydro Aluminium, Sunndal Verk

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Foreword

Prof. Altenpohl's book, "Aluminum Viewed from Within", bridges a gap between physical metallurgy and industrial application of aluminum. Already four editions were printed in German and translations into Japanese and French have been published. There is no other book on the metallurgy of aluminum and its alloys, having such a wide distribution.

Besides its usefulness for students, the main purpose of the book is to arouse the interest of people employed in the aluminum industry to achieve a better understanding of manufacturing processes of the metal and its alloys through a step-by-step explanation. This is accomplished by describing transformations in the metallic structure like the age hardening process, and by providing a certain understanding of metal physics. There is much emphasis placed on the knowledge of structural changes which take place during fabrication of aluminum and its alloys, from the raw material to the finished product. The casting technique is significant not only for the properties of castings but also how the cast structure affects the properties of rolled or extruded products. The steps involved in the fabrication process are divided into mechanical and thermal treatments. In every case, the changes in the properties are based on transformations within the atomic structure of the material.

"Aluminum Viewed from Within" applies to the practical man in the plant, providing a path to metallurgical understanding. The practitioner has no trouble following the subject because the information is presented simply and requires very little previous knowledge of the subject. The book is no less useful to the designer to whom an insight into the specific properties of aluminum and its alloys is indispensable. This book, by the way, is an off-spring of a large volume "Aluminium and Aluminium Alloys", written by the author together with other reknown experts and published by Springer Verlag in 1965. It combines results of classical metallurgy, metal physics and materials technology.

It wish the English edition the same success as the German, Japanese and French editions.

Professor Werner Koester, PhD,
Hon.-Dr. Eng., Director Emeritus,
Max-Planck Institute for Metal Research,
Stuttgart, Germany.

Figure 148d shows the structure of an extruded rod. By far, the majority of the cross-section consists of a fibrous structure, with only the surface showing a thin recrystallized zone. A medium to coarse grain on the surface of extruded shapes cannot be prevented for some alloys, but for the most part its occurence is tolerable, since the stronger fiber structure occupies the greater part of the cross-section. But, the recrystallized surface of the extrusion may be undesirable for decorative applications (increased work required for polishing to avoid streaking).

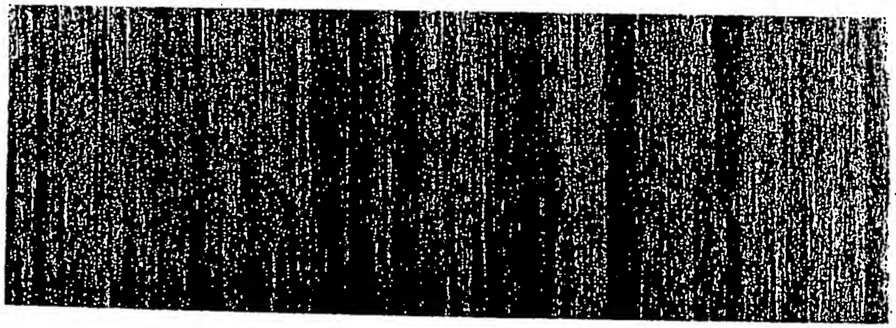

Figure 149: Soft, commercial purity aluminum sheet (etched) with a fine-grained but streaked structure. The streaks are due to large as-cast grains, which become evident again after recrystallizing twice. This streakiness becomes visible after etching or anodizing.

Causes of Streaking or Streak-Free Structure after Forming

The severe reduction in cross-sectional area of 50 : 1 to 80 : 1 during extrusion produces a more uniform structure than is possible in sheet, on which a streaked structure often appears during anodizing (Figure 149). For various reasons it is undesirable to have either a hot-rolled structure which contains non-recrystallized grains from the cast structure, or one with coarse recrystallized grains. The outline of large crystals still present after hot deformation can often be recongized after subsequent cold working (for example, after anodizing, it shows up as a streaked structure).

The streaking from large as-cast grains even after severe cold work is not hard to understand. The as-cast grains are surrounded by heterogeneities. The grain boundary material is not displaced during recrystallization, and is usually elongated only in one direction during subsequent cold work. The localized enrichment of inclusions can be recognized after etching, polishing or anodizing, of sheets or shapes, as a streaked structure, since the inclusions usually are attacked either more or less than the matrix by chemicals and they discolor the anodic film (Figure 149).

In addition to the size of the as-cast grains, their cell size is of importance for the streakiness of the sheet. Large-celled structures or a large variation in the cell size are undesirable (see Figure 39 on page 55). With today's improved DC casting techniques, it is possible to produce rolling ingots with a fine-celled, fine-grained and uniform as-cast structure which produces streak-free sheets.

The situation is similar for coarse grains in wrought material. After sufficient cold work and recrystallization, the original large grains will produce small crystals with similar orientation, regardless of whether the original large grains were as-cast or the result of recrystallization. Thus, even though the final strucure is fine-grained, the original "mother crystal" is recognizable.

It is therefore important to avoid coarse grain in all processing steps, including casting, hot working and intermediate annealing, in order to obtain a finished product with the most uniform surface and forming characteristics possible (quasi-isotropic state). Modern fabrication methods fulfill these requirements for all wrought alloys, with few exceptions.

Furnaces

So far, the fundamental aspects for thermal treatment of aluminum and aluminum alloys have been discussed. It should be kept in mind that heat-treating operations for aluminum are precision processes. Therefore, the furnaces in which the thermal processes are carried out must be designed and maintained properly to ensure reproducibility and uniformity of time-temperature cycles.

The effects of time and temperature, together with the necessity to conduct many treatments at temperatures near the eutectic melting point of the alloy makes close temperature control of an entire furnace charge mandatory during the heat-treatment cycle. Whenever possible load-thermocouples should be used for direct temperature measurement.

Generally, standard types of furnaces and heating equipment can be used for heat-treating aluminum alloys. These include car and truck type, vertical pit and tower, horizontal conveyor, and strip- and sheet-processing furnaces.

The selection of furnace equipment depends on process and product requirements. In practice this means the choice between batch and continuous operation.

Batch furnaces usually permit a greater load density than continuous furnaces. Normally batch furnaces are used for products which are massive and heat slowly, and processes which involve a soak (holding at temperature) so that the heat-treatment cycle is long regardless of the time required to reach temperature. Typically, forgings, cast and extruded parts are handled in batch furnaces.

Batch thermal treatment of coils of wire and sheet present special problems. Coils of these products have poor thermal conductivity in the radial direction which means that they have a much slower heating and cooling rate inside, than at the surface. Therefore, quenching and precipitation treatments are generally not possible. With increasing coil weight (up to 10–20 tons for coils of sheet), variations in strength and grain size become more evident after partial annealing and even full annealing. The very long thermal cycles of more than 24 hours also result in low furnace throughput. Continuous furnaces provide a solution to these problems.